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СОРБЦИЯ ТЯЖЕЛЫХ МЕТАЛЛОВ МНОГОКОМПОНЕНТНЫМИ ПЕНОПОЛИМЕРНЫМИ СОРБЕНТАМИ

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Статья посвящена исследованию процесса сорбции тяжелых металлов из водной среды пенополимерными сорбентами, полученными на основе многокомпонентных полимерных смесей. Потребность в таких исследованиях продиктована необходимостью получения полимерных сорбентов с различными функциональными или полярным группами, фактически способствующими значительному увеличению эффективности сорбции тяжелых металлов пенополимерными сорбентами. Бикомпонентные полимерные смеси, полученные на основе полиамид + полиуретан, а также на основе трехкомпонентной смеси полиамид + полиуретан + сополимер бутадиена, стирола с акрилонитрилом, использовались в качестве полимерной матрицы пенополимерных сорбентов с различным соотношением полярных групп. В зависимости от химической природы и соотношения компонентов смеси на основе полиуретана, полиамида и сополимера бутадиена со стиолом и акрилонитрилом, в дополнение к группам уретана, эфира, амида и мочевины в составе композиции зафиксированы такие функциональные группы, как алифатические и нитрильные группы. Приводятся результаты исследования кинетической закономерности сорбции тяжелых металлов из водной среды в зависимости от типа и соотношения полимерных компонентов в составе сорбента. Показано, что структура полярного пенополимерного сорбента состоит из пор и ячеек с мембранной структурой, позволяющей использовать это обстоятельство для эффективной сорбции тяжелых металлов. Концентрация сорбированного тяжелого металла в составе сорбента была определена по разработанной методике УФ- спектрального анализа. Показано, что наличие системы пор и ячеек в структуре пенополимерного сорбента способствует протеканию сорбции в наиболее глубоко расположенных его участках. Это обстоятельство дает основание для утверждения о том, что в данном случае имеет место объемная сорбция тяжелых металлов. По существу, структура сорбентов пенополимера способствует концентрации сорбата на поверхности их мембран. Большое разнообразие адсорбционных центров на поверхности и по всему объему сорбента способствует протеканию целого комплекса взаимодействий сорбент-сорбат, среди которых взаимодействия Ван-дер-Ваальсовских сил, электростатического взаимодействия и водородных связей – являются главными.

Ключевые слова: пенополимерный сорбент, объемная масса, сорбционная способность, макроструктура

SORPTION OF HEAVY METALS BY MULTICOMPONENT FOAM POLYMER SORBENTS

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The article is dedicated to the investigation of the sorption of heavy metals from water bodies by foam polymer sorbents obtained on the basis of multicomponent polymer mixtures. The need for such studies was dictated by the fact that the more diverse the content of functional or polar groups became in the composition of the foam polymer sorbent, the greater was the probability of increasing the efficiency of sorption of heavy metals. Biocomponent polymer mixtures based on polyamide + polyurethane and three-component mixtures polyamide + polyurethane + acrylonitrile-butadiene-styrene copolymer were used as polymer matrix of the foam polymer sorbents at various ratios. Depending on the chemical nature and the ratio of the initial components in the macromolecules of polyurethane, polyamide and acrylonitrile-butadiene-styrene copolymer, in addition to urethane groups, functional groups such as ether–, ester, amide, urea, and also aliphatic and nitrile groups are also present. The investigation results of the kinetic regularities of heavy metals sorption under polymer components different ratios and ambient temperature are presented. It is established that the foam polymer sorbent consists of pores and cells with a membrane structure, which allows using it for effective sorption of polar and nonpolar compounds. The concentration of sorbed heavy metals was determined by UV-spectroscopic analysis. It is shown that the presence of a system of pores and cells allows the sorbate to diffuse into its more deeply located regions, as a result of which sorption occurs throughout the volume of the sorbent. Essentially, the foam polymer sorbents sorb the molecules by dissolving them on the surface of their membranes. A large variety of adsorption centers on the surface and in the bulk of the foam polymer causes a whole complex of sorbent-sorbate interactions, among which the Van der Waals interactions, dispersion interactions, electrostatic interactions and hydrogen bonds are the main ones.

Key words: foam polymer sorbent, bulk weight, sorption capacity, macro structure

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INTRODUCTION

Constant and growing environmental pollution by hydrocarbons and heavy metals occurs due to development and expansion of different industries [1-7]. At the same time, not only the soil and water bodies are polluted, but also the air environment, which, as is known, adversely affects the environment and human health. At this stage, we are going through a rather difficult period in our activity, and therefore it becomes necessary to carry out a set of measures aimed at eliminating or neutralizing centers that are dangerous to the health of people and their descendants. At present, water quality in natural sources has deteriorated significantly. A special group presents a number of heavy metals, many of which are toxic.

A weighty argument in favor of purifying the natural water environment is the degree of influence of metals on human health. There are metals that even with a slight ingress into the human body cause irreparable harm to his health with a possible fatal outcome. At the same time, there are metals that are constituent part of the body, performing a useful and necessary mission under certain micro doses to strengthen human immunity. If in the human body, the content of copper, cobalt or chromium ions does not exceed the required dose, then they can be considered as trace elements. At their considerable excess in the body, the term "heavy metals" comes into full force.Therefore, it will be possible to agree with the opinion that there are no harmful substances, but there are dangerous concentrations for human health.

In recent years, the interest of scientists in the development of sorbents for the selective sorption of those or other metal ions from the aqueous medium has significantly increased [8-13]. A relatively new sorbent for the sorption of heavy metals is the foam polymer materials. This is explained, first of all, by the relatively large sorption capacity of foam polymers for hydrocarbons and heavy metals, their buoyancy and the ability to repeatedly regenerate. Unexplored in terms of use for cleaning reservoirs from heavy metals are foam polymer sorbents, especially derived from multicomponent polymer blends.

In this regard, the purpose of this paper is to conduct researches aimed at studying the sorption characteristics of multicomponent foam polymer sorbents for various types of heavy metals.

EXPERIMENTAL

The foam polymers, which developed and obtained by us on the basis of polymer mixtures have been used as the object of the research. The initial components of the polymer mixture were polyurethane (PU), polyamide (PA) and acrylonitrile-butadiene-styrene copolymer (ABS). Foam polymer sorbents were being obtained during the mixing of polymer components in the melt regime, into composition which the dicumyl peroxide and porophore were simultaneously added. The mixing process was being carried out in the material cylinder of the extrusion machine [14-16].

The sorption properties of the foam polymer sorbents have been studied in aqueous solutionof metal salts. The change in the concentration of heavy metals were being periodically measured by spectrophotometry method, by which were being determined the kinetic regularities of their sorption.

Chemical modification of the foam polymer sorbents was being carried out as follows: To remove air bubbles from the sorbent cells, the crumb or granules were pushed by glass rod.The granules were mixed with a vibro-mixer in a vessel containing sodium nitrite (at the rate of 2.4 g/l) in 30 ml of 2 M aqueous solution of hydrochloric acid.

RESULTS AND DISCUSSION

Depending on the chemical nature and the ratio of the initial components in the macromolecules of PU, PA and ABS-copolymer, the following functional groups are presented: urethane, ether -O-, ester-C(O) -O-, amide -C(O)-NH-, urea NH_2-C(O)-NH_2, and also aliphatic -CH_2- and CN-groups.

Foam polymer sorbent on the basis of PU, PA and ABS consists of pores and cells with a membrane structure, which allows using it for effective sorption of polar and nonpolar compounds. The presence of the pore system and cells allows diffusion of sorbate in its more deep-seated areas. Unlike polymer plates in our foam polymer sorbents sorption occurs throughout the volume of the sorbent. In this case, the extraction of compounds occurs not only due to adsorption (surface adsorption), but also as a result of absorption (absorption by all volume of polymer). Essentially, the foam polymer sorbents sorb the molecules by dissolving them on the surface of own membranes. A large variety of adsorption centers on the surface and in the volume of polymer PU (PPU) causes a whole complex of sorbent-sorbate interactions, among which Van-der-Waals, dispersion, electrostatic and hydrogen bonds are the main ones. The role of each of these types of bonds largely depends on the structure of the polymer link and the chemical nature of the molecules being sorbed [17-19].

In order to show how effective the foam polymer sorbents are for sorption of heavy metals, let us turn to the results of the study given in Table.

Comparative analysis of the data unequivocally proves the extent to which foam polymer sorbents have wide possibilities. Table 1 gives data on sorbates based on ions of heavy metals such as Cu^{2+}, Co^{2+} and Ni^{2+}. Analyzing the data, it can be found that the foam polymer sorbents on the basis of PA + PU + ABS have comparatively high sorption characteristics. As can be seen from this table, the use of foam polymer sorbent modified with sodium nitrite ($NaNO_2$) contributes to a significant increase (in 2-2.5 times) in sorption capacity for heavy metals. To obtain more detailed information on the sorption capabilities of polypropylene sorbents based on multicomponent polymer mixtures, we turn to a phased analysis of the results of the study. Fig. 1 shows the sorption curves, obtained out at different temperatures, of copper ions on polyurethane sorbents based on a mixture (PU + PA), which differ in the ratio of the components of the mixture. From a comparative analysis of the data, it can be established that the highest values of sorption of copper ions are observed in the range of the ratios PU: PA = 60:40 - 40:60. Moreover, with increase in the temperature of the aqueous medium from 298 to 318 K, the sorption capacity of the copper ion increases.

Table

Physicochemical and analytical characteristics of the foam polymer sorbents at sorption of Cu^{2+}, Cd^{2+}, Ni^{2+} under 298 K. The bulk weight of the sorbent is 200-300 kg/m^3. Time is 20 min

Таблица. **Физико-химические и аналитические характеристики пенополимерных сорбентов при сорбции Cu^{2+}, Cd^{2+}, Ni^{2+} при температуре 298 К. Объемная масса сорбента 200-300 кг/м^3. Время 20 мин**

№	Sorbent composition	Sorbate, Me^{2+}	pH	Sorption capacity for Me^{2+}, mg/g
1	PA+50%PU	Cu^{2+} Cd^{2+} Ni^{2+}	3.5-4.5 4.0-4.5 4.0-5.0	4.2 4.5 4.8
2	(PA+50%PU)+ 10%ABC	Cu^{2+} Cd^{2+} Ni^{2+}	3.5-4.5 3.5-4.5 4.5-5.0	5.3 4.9 5.5
3	Modified (PA+50%PU)+ 10%ABC	Cu^{2+} Cd^{2+} Ni^{2+}	3.0-4.5 3.5-4.0 3.5-4.5	12.6 13.3 11.2

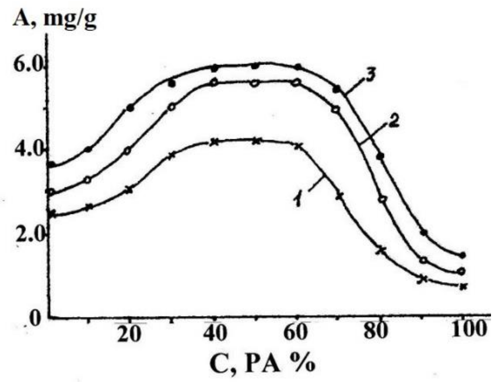

Fig. 1. Effect of PA concentration in the composition of the foam polymer sorbent on the basis of PU + PA and the temperature of the aqueous medium on the sorption capacity for Cu^{2+} ions: 1- 298 K; 2- 308 K; 3 - 318 K

Рис. 1. Влияние температуры водной среды и концентрации ПА в составе пенополимерного сорбента на основе ПУ+ПА на сорбционную емкость по иону Cu^{2+}: 1- 298 К; 2- 308 К; 3- 318 К

Taking into account the fact that the composition of the sorbent mixture materially affects the sorption capacity, it seemed interesting to study the effect of the ABS-copolymer on the regularity of the change in the value of this parameter. Analyzing the data presented in fig. 2, one can note that with the introduction of ABS-copolymer into the PU + PA mixture, there is a general tendency towards an increase in the sorption capacity of the foam polymer sorbents obtained on their basis. At the same time, the growth of the sorption capacity for copper ions proceeds throughout the entire range of PU + PA ratios.

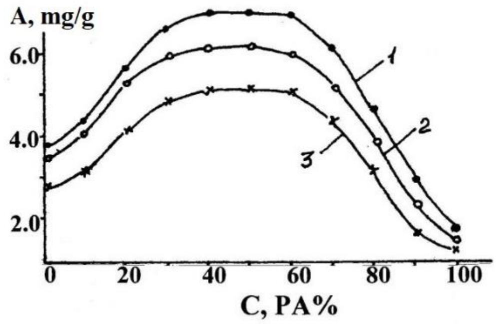

Fig. 2. Effect of PA concentration in the composition of the foam polymer sorbent on the basis of (PU + PA) + 10% wt. ABS and the temperature of the aqueous medium on the sorption capacity for Cu^{2+} ions: 1- 318 K; 2- 308 K; 3- 298 K

Рис. 2. Влияние концентрации ПА в составе пенополимерного сорбента на основе (ПУ+ПА)+10%масс.АБС и температуры водной среды на сорбционную емкость по ионам Cu^{2+}: 1- 318 К; 2- 308 К; 3- 298 К

Fig. 3 shows the kinetic regularity of copper ions sorption for foam polymer sorbents based on individual polymers of PPU and PPA, as well as their mixtures. As can be seen from Fig. 3, the sorbents based on PPA, and then polyurethane foam, possess the lowest sorption capacity. At the same time, the foam polymer sorbents based on mixture of polymers are characterized by higher values of the sorption capacity for copper ions. The obtained experimental results of the study allow to consider that the additional introduction of ABS copolymer into the the polymer mixture PA + PU makes it possible to some extent to increase the sorption capacity. Apparently, this is due to the fact that in addition to the available active functional groups [ether -O-, ester -C(O)-O-, amide -C(O)-NH-, urea NH_2-C(O)-NH_2], CN- groups are added that can further activate all available polar groups to sorption of heavy metals.

The presence of end amine group in the composition of PA and PU opens up a promising possibility of modifying their molecular structure. As a result, it is possible to significantly increase the sorption capacity of the foam polymer sorbents. As a result of modification of the terminal amine groups in the composition of PPU and PPA, it is possible to obtain a polymer cation of diazonium according to the known scheme:

$$RNH_2 \xrightarrow[2M\ HCl]{NaNO_2} R\text{-}N^+ \equiv NCl^-$$

As can be seen from the presented chemical reaction, a cation of diazonium is formed in the hydrochloric acid aqueous solution of sodium nitrite, which, as is well known, is characterized by high activity with respect to heavy metal ions. Evidence of this was the results of IR spectral analysis of diazotized PPU and PPA, according to which absorption bands in the region of 2106-2110 cm^{-1} appeared on the spectra of these modified polymers. Their appearance is usually interpreted by the vibrations of the N≡N bond, which indicates the formation of the diazonium salt in the polymer phase. Simultaneously, a redistribution of the intensity in a wide band of NH_2-valent vibrations was observed in the 3260-3410 cm^{-1} region, which indicates the diazotization reaction involving NH_2 groups [5, 20-21].

Comparing the results of the study in Figs. 1 and 4, it can be established that the process of copper ions sorption on the modified sorbent in the entire range of the PU:PA ratios proceed more efficiently. In this case, the sorption process on sorbents is most effective in the range of the ratio PU: PA = 70:30 - 40:60. With increase in the temperature of the aqueous medium from 298 to 318 K, increase in the rate of sorption and sorption capacity is observed. All these circumstances indicate that chemisorption occurs on the surface of the membranes of the foamed polymeric sorbent. Effective stirring of the sorbent in the solution allows to maintain a constant concentration of the sorbed dissolved copper ion near the sorbent-sorbate interface and thus to exclude the influence of the diffusion process from the volume of the solution on sorption.

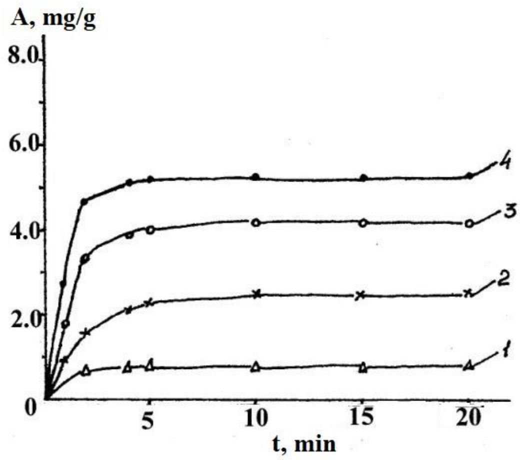

Fig. 3. Kinetic regularity of sorption of Cu^{2+} by foam polymer sorbents with bulk weight of 50-70 kg/m^3: 1-PA; 2- PU; 3- PA + 50% wt of PU; 4- (PA + 50% wt PU) + 10% wt of ABS

Рис. 3. Кинетическая закономерность сорбции Cu^{2+} пенополимерными сорбентами с объемной массой 50-70 кг/м^3: 1- ПА; 2- ПУ; 3- ПА+50% масс.ПУ; 4- (ПА+50%масс.ПУ)+10%масс.АБС

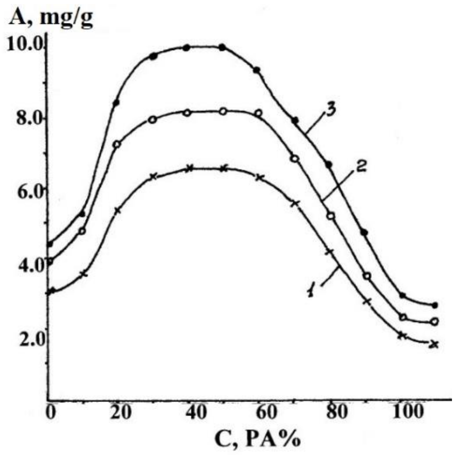

Fig. 4. Influence of temperature and concentration of PA in the composition of chemically modified foam polymer sorbent (PA + 50% wt of PU) + 10% wt of ABS on the sorption capacity for copper ions: 1- 298 K; 2- 308 K; 3 318 K. The bulk weight of the sorbent is 50-70 kg/m^3

Рис. 4. Влияние температуры и концентрации ПА в составе химически модифицированного пенополимерного сорбента (ПА+50% масс.ПУ) +10% масс.АБС на сорбционную емкость по ионам меди: 1- 298 К; 2- 308 К; 3- 318 К. Объемная масса сорбента 50-70 кг/м^3

The obtained results of the study are in good agreement with the previously developed ideas on the possibility of chemical modification of the polyurethane foam. In fact, we have the opportunity to obtain new materials – chemically modified polyurethane foam. Assuming that the diazonium salts can have a sufficiently high activity with respect to the salts of heavy metals, it seemed interesting to study the sorption properties of modified polymeric foam sorbents obtained on the basis of a mixture of PA + PU + ABS. For example, copper salts in an aqueous medium with concentration of 300 mg/l in the initial mixture have been used as the sorbate. Fig. 4 shows the results of a study of the effect of aqueous medium temperature on the sorption capacity of a chemically modified expanded polymer sorbent obtained on the basis of a mixture of PU + PA + ABS.

In those cases, when the foam polymers are used as a sorbent, the important moment is to study the influence of their bulk weight on sorption processes. So, for example, we used as the object of research foam polymer sorbents on the basis of a mixture of PU + PA + ABS with different bulk density and, respectively, cell diameter (Fig. 5).

In this case, we used 3 types of sorbent with bulk density of 50-70, 200-300 and 500-600 kg/m^3, respectively, with cell diameter equal to 0.9-1.1, 0.1-0.2 and 0.01-0.02 mm. In addition, a modified sorbent with bulk density of 200-300 kg/m^3 has been used. Comparing the curves in this Fig. 5, one can pay attention to the fact that, with other equal conditions, sorbents with bulk weight of 200-300 kg/m^3 (curve 1) possess comparatively better sorption properties. Worst sorption properties have sorbents with bulk density of 500-600 kg/m^3 (curve 3). It is possible that in this case, it would be appropriate to mention the peculiar "capillary" effect that occurs in porous or close-cell macrostructures of sorbents.

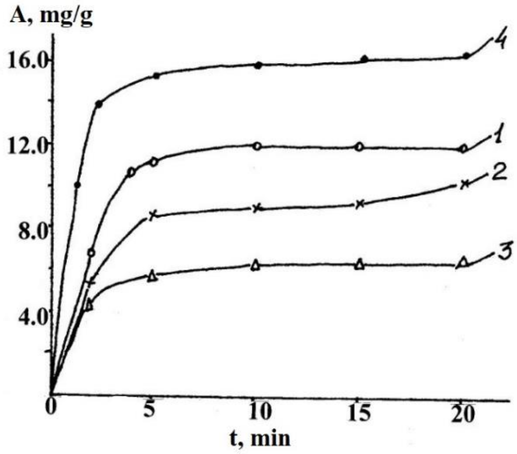

Fig. 5. Kinetic regularities of Cu^{2+} sorption by chemically modified foam polymer sorbents on the basis of (PA + 50% wt PU) + 10% wt of ABS with different bulk weight: 1- 200-300 kg/m^3; 2- 50-70 kg/m^3; 3- 500-600 kg/m^3; 4- chemically modified sorbent with a bulk weight of 200-300 kg/m^3. The temperature of the aqueous medium is 318 K

Рис. 5. Кинетические закономерности сорбции Cu^{2+} химически модифицированными пенополимерными сорбентами на основе (ПА+50%масс.ПУ)+10%масс.АБС с различной объемной массой: 1- 200-300 кг/м^3; 2- 50-70 кг/м^3; 3- 500-600 кг/м^3; 4- химически модифицированный сорбент с объемной массой 200-300 кг/м^3. Температура водной среды 318 К

According to this theory, as the diameter of the cells decreases or the foam is compacted, the size of the cells decreases to such an extent that it favors the development of a dispersion interaction between the sorbate and the functional groups of the sorbent. As a result, the sorption capacity and the degree of extraction of metal ions increase. But with the further consolidation of the foam polymer sorbent within 500-600 kg/m^3 and higher, the diameter of the cells sharply decreases to 0.01-0.02 mm, which immediately affects on the one hand, on diffusion difficulties in the delivery of metal ions to deeper sections of the sorbent, and with another, the difficulty of removing air from the fine-mesh structures of sorbents. As a result, the sorption of metal ions in fine-grained macrostructures with a bulk density of 500 kg/m^3 and above becomes inefficient, since it proceeds mainly on the surface areas. As for sorbents with a bulk density of 50-70 kg/m^3, in this case, in connection with the increase in the cell volume, we are already confronted with the opposite effect: the possible weakening of the dispersion forces of interaction between sorbent functional groups and metal ions. As can be seen from Fig. 5 (curve 2), this circumstance to a certain extent leads to a decrease in the degree of recovery of the sorbate.

If the sorbent with a bulk density of 200-300 kg/m^3 is subjected to chemical modification it can be seen that the sorption capacity of these samples rises sharply from 12.0 to 16.0 mg/g (Fig. 5, curve 4). The obtained results of the study once again confirm the high efficiency of the diazonium salt during the sorption of copper ions.

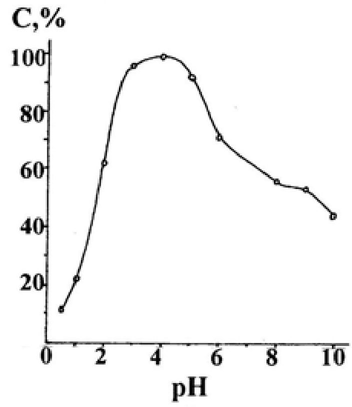

Fig. 6. Influence of the pH on the degree of extraction of Cu^{2+} from the aqueous medium by the foam polymer sorbents on the basis of modified (PA + 50% wt PU) + 10% wt of ABS. The volume weight of the sorbent is 200-300 kg/m^3

Рис. 6. Влияние pH среды на степень извлечения Cu^{2+} из водной среды пенополимерными сорбентам на основе модифицированного (ПА+50% масс.ПУ) +10% масс. АБС. Объемная масса сорбента 200-300 кг/м^3

In order to obtain a more complete picture of the processes that occur at the sorbent-sorbate interface, it seemed interesting to study the effect of pH of the medium and the bulk density of the foam polymer sorbent on the sorption efficiency. Fig. 6 shows the study results of the extraction degree of copper ions by multicomponent and chemically modified foam polymer sorbents on the basis of a mixture of PU + PA + ABS. From the analysis of the curves in this figure, it can be established that comparatively better sorption occurs in media where the pH of the medium has values in the range of 3-5.

CONCLUSION

Based on the studies conducted, it can be argued that polymer blends based on PA + PU + ABS are promising polymer composites for the preparation on their basis of hydrophilic foam polymer sorbents intended for selective sorption of heavy metals from an aqueous medium.The more different types of functional groups in the composition of the foam polymer sorbent, the more active centers for the effective course of sorption of heavy metals. It becomes obvious that the use of foam polymer sorbents on the basis of individual polymers can not ensure their high sorption capacity.

Along with this, carrying out of chemical modification of foam polymer materials with sodium nitrite on the basis of terminal amine groups present in the composition of PU and PA allows to get even more active centers - diazonium cations, which resultsin the maximum effect on improving sorption capacity of foam polymer sorbents.

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